AEROSOL PROPERTIES AND PROCESSES: A PATH FROM FIELD AND LABORATORY MEASUREMENTS TO GLOBAL CLIMATE MODELS

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Aerosol Properties and Processes: A Path from Field and Laboratory Measurements to Global Climate Models

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Abstract

    Aerosols in the lower atmosphere exert a substantial influence on climate and climate change through a variety of complex mechanisms. Consequently there is a need to represent aerosol effects in global climate models, and models have begun to include representations of these effects. However, the treatment of aerosols in current global climate models is presently highly simplified, omitting many important processes and feedbacks. Consequently there is need for substantial improvement. Here we describe the U. S. Department of Energy strategy for improving the treatment of aerosol properties and processes in global climate models. The strategy begins with a foundation of field and laboratory measurements that provide the basis for modules of selected aerosol properties and processes. These modules are then integrated in regional aerosol models, which are evaluated by comparing with field measurements. Issues of scale are then addressed so that the modules can be applied to global aerosol models, which are evaluated by comparing with global satellite measurements. Finally, the validated set of modules is applied to global climate models for multi-century simulations. This strategy can be applied to successive generations of global climate models.

The challenge. Aerosols influence climate by modifying the global energy balance through absorption and scattering of radiation (direct effects) and by modifying cloud reflectance and persistence, modification of precipitation (indirect effects). Quantification of the influences of aerosols, and of anthropogenic aerosols, on Earth's radiation budget is of high priority for relating historical climate change to increasing greenhouse gases and determining Earth's climate sensitivity, as the uncertainty in the radiative forcing by aerosols greatly exceeds that of all other forcing mechanisms combined. Aerosols contribute to numerous other climatically important processes, including fertilization of land and oceans through deposition of nitrate, iron, and other nutrients, acidification of lakes and forests through deposition of sulfate, and reduction of snow and ice albedo through deposition of black carbon. Although field measurements and remote sensing by satellites provide essential information regarding aerosol loading, geographical and vertical distribution, processes and influences, such measurements are necessarily limited in space and time. Hence accurate assessment of aerosol influences climate simulations by numerical models is essential for interpreting past climate and for examining future changes for various emission scenarios. This assessment requires accurate representation of aerosol physical and chemical properties and of the processes that influence those properties within global climate models.

The requirement. The climate influences of anthropogenic aerosols are very difficult to determine separately from the effects of natural aerosols using observations alone. Hence these influences are most effectively determined from climate simulations carried out with and without anthropogenic aerosols. Increasingly it has become recognized that aerosols are highly interactive with other components of the climate system, for example influencing and being influenced by clouds and precipitation. This recognition leads to a requirement for climate models that intrinsically incorporate representation of the amount and properties of aerosols as a function of three-dimensional location and time. Such a representation in turn requires understanding of the governing processes and the dependence of these processes on amounts and properties of particulate matter and gaseous precursors. Aerosol direct effects on the energy balance of the Earth depend on the optical properties of the aerosol, specifically the extinction coefficient σ_{ep}, the single scattering albedo ω_0 (fraction of extinction coefficient due to scattering), and asymmetry parameter g (mean of cosine of scattering angle). Aerosol indirect effects depend on the number concentration of cloud condensation nuclei (CCN) as a function of supersaturation s, $N_{CCN}(s)$. These aerosol properties in principle are evaluated as integrals over the properties of the individual particles that comprise the aerosol. The single-particle properties are governed by the particle size and shape, by the properties of the different materials the particle is composed of, and by the distribution of the materials within the particle. However, as representation of these aerosol properties in such detail is beyond the capability of present climate models as well as those of the foreseeable future, the challenge of climate modeling is to represent this complexity within the constraint of computational resources. This requires compromises and assumptions that accurately account for the most important aerosol effects within the constraints of practical application.

Aerosol properties that must be accurately represented include mass concentration, particle size and size-dependent composition, optical properties, solubility, and ability to serve as nuclei of cloud particles. Key processes (Figure 1) which must be represented include emissions of

primary particles (those emitted directly into the atmosphere) such as mineral dust, sea salt, black carbon, and organic carbon; emissions of aerosol precursor gases such as dimethylsulfide (DMS; all acronyms are listed in the appendix), sulfur dioxide, and volatile organic compounds (VOC); oxidation of precursor gases, new particle formation, reversible growth of particles by condensation of nonvolatile and semivolatile gases, coagulation, reversible uptake of water vapor, activation to form cloud droplets, in-cloud scavenging, aqueous-phase reactions in cloud droplets, subgrid vertical transport by clouds, cloud-drop evaporation, dry deposition to the surface, and scavenging by falling hydrometeors. Accurate representation of these processes in climate models rests not only on understanding of the pertinent processes but also on development of efficient ways of representing them in large-scale numerical models.

A Strategy. As the treatment of aerosol properties and processes in present global climate models (GCMs) is highly simplified, opportunities for improving the treatment abound. Four stages in a possible strategy to represent aerosol processes in climate models and examine aerosol influences on climate are summarized in Table 1. Stage 1 focuses on improving understanding of isolated processes. Stage 2 develops and evaluates modules representing these processes. Stage 3 evaluates the interaction of those modules in integrated aerosol models. Once validated, the modules are incorporated within global climate models in stage 4 and again evaluated. At the end of this process the models are available to examine the sensitivity of climate to different emissions scenarios.

Here we describe the strategy (Figure 2) for improving the representation of aerosol properties and processes in GCMs that is being executed by the U.S. Department of Energy (DOE) as part of its contribution to improving representation of climate in large scale numerical models. The U.S. DOE supports three research programs that contribute to the improvement of the representation of aerosol properties and processes in GCMs: the Atmospheric Science Program (ASP), the Atmospheric Radiation Measurement (ARM) Program, and the Climate Change Prediction Program (CCPP). Each program plays a vital role in the DOE strategy to improve the accuracy and predictive capability of large scale climate models. The ASP and ARM provide field and laboratory measurements that guide the development of models of specific aerosol properties and processes. The ASP has an explicit charge to improve understanding of chemical, microphysical, optical, and cloud nucleating properties of aerosols and of the processes that control those properties and to develop numerical models of these processes suitable for inclusion in large-scale models. ARM has a broad mission of understanding and quantifying the atmospheric processes and properties that influence radiation, including both direct and indirect effects aerosols, and of developing and evaluating process models. The CCPP supports the application of process models as modules in GCMs so that the influence of anthropogenic aerosols on climate can be quantified in the context of climate change over the industrial period.

This DOE strategy is embedded within the larger context of the U. S. Climate Change Science Program, the World Climate Research Program (WCRP) and the International Geosphere-Biosphere Program (IGBP). The U. S. National Aeronautics and Space Administration, the National Science Foundation, the National Oceanic and Atmospheric Administration, the U. S. Environmental Protection Agency, the WCRP, and the IGBP all provide important contributions to such a strategy. Elements of this overarching strategy have been described by Diner et al. (2004) and Ackerman et al. (2004).

Generations of Climate Models. Climate model studies over the past ten years have shown that aerosol forcing must be represented in climate models in order for these models to accurately represent temperature change over the industrial period. Early approaches to such representation simply adjusted surface albedo to crudely account for the enhancement of planetary albedo due to tropospheric aerosols (Mitchell et al., 1995, Boer et al., 2000; Delworth and Knutson., 2000). Subsequently, many models have treated direct effects of aerosols using three-dimensional distributions from "offline" simulations with chemical transport models (e.g., Meehl et al., 2003, Delworth et al, 2006; Collins et al., 2006; Meehl et al., 2006). Some GCMs have actively represented certain aerosol components (usually sulfate) and processes (e.g., Stott et al., 2000, Tett et al., 2002; Hansen et al., 2005; Schmidt et al., 2006). It is now recognized that accurate representation of aerosol influences must take into account such phenomena as correlations of aerosol loading with meteorological variables and the influences of aerosols on clouds and precipitation and hence that aerosol loading and properties must be represented actively and interactively in climate models. It is this recognition that is driving much of the current effort to actively represent aerosol processes, properties, and effects in climate models.

Much of the practice of climate modeling has become tied to the production schedules for periodic international assessments of the science of climate change by the Intergovernmental Panel on Climate Change (IPCC). The fourth IPCC assessment report (AR4), to be published in 2007, will be based on simulations completed in 2004 using models that were frozen in 2003. As these assessments are prepared every roughly six years, it may be anticipated that the fifth IPCC assessment (AR5) will be based on climate simulations completed in about 2010 using models frozen about 2009. Likewise the sixth assessment (AR6) will be based on models frozen roughly in 2015. Each IPCC assessment report is therefore based on a successive generation of climate models, with the timing of model development and application to some extent governed by the timing of the assessment process. Although climate modeling predates IPCC assessments by many years, recent generations of climate models can be identified by the IPCC report to which they contribute. Thus, we might say that the fourth generation of climate models contributed to the preparation of AR4, and so on.

Inevitably, there is a lag from understanding of processes to their representation in climate models, and this holds true for aerosol processes. Thus there is a lag from understanding of aerosol processes as represented in zero-dimensional models (box models) to representing this understanding in integrated aerosol models, and a further lag in representing this understanding in GCMs that are used in IPCC assessments; this lag can be as long as a full IPCC cycle or more. There is a further, similar lag of a full IPCC cycle between the representation of aerosol (or other) processes in GCMs and the use of the results of that generation of models in scenario assessments.

Treatment of aerosols in future generations of climate models will rest on improved understanding of the processes that control aerosol properties and their evolution as gained in laboratory and field studies carried out in research supported by the DOE ASP and ARM programs, as well as by studies conducted with support of other U.S. and international agencies. These studies in turn rest on developments in instruments and measurement capabilities to characterize aerosol properties.

Future enhancements will rest also on advances in representing the pertinent aerosol processes in models. Such model development progresses through a hierarchy of approaches. Initially a subset of aerosol processes is represented in zero dimensions or, for cloud processes, in one (vertical) dimension. These representations are then commonly incorporated into models that are driven by analyzed meteorological data to allow the representation of these processes to be evaluated by comparing with observations at specific locations and times. These process models subsequently become incorporated as modules in integrated aerosol models that are further tested and evaluated. Ultimately these representations become incorporated in climate models. As climate models must be run for much greater times (centuries) than is typical for aerosol models, the ability to include aerosol processes will require accurate efficient representations of aerosol processes and will rest as well on advances in computational hardware and architecture. It is thus clear that representing aerosol influences on climate, at present and for the foreseeable future, will require a suite of approaches. Examples of these approaches are given in Box 1.

The incorporation of these approaches in present and future generations of climate models is outlined below, together with examples of research needed to support development and testing of these new approaches. Aerosol properties that need to be represented in climate models to simulate aerosol influences on climate are presented in Table 2 together with approaches to modeling these properties in the several model generations. The approaches representing aerosol processes are given in Table 3.

Box 1. Multiple approaches for multiple applications. Aerosol models :

- May examine individual processes or interactions of multiple processes.

- May serve as testbeds for new representations of aerosol processes in climate models.

- May be driven by analyzed meteorological data (offline), neglecting aerosol influences on meteorology and climate.

- May produce aerosol distributions that are prescribed in climate simulations.

- May include representation of aerosol influences on meteorology and climate (aerosol feedback).

- May include explicitly resolved or parameterized clouds.

- May examine the consequences of alternative emissions scenarios.

Aerosols in Fourth Generation Climate Models. The summaries of aerosol properties and processes included in fourth generation climate models, frozen in 2003 or early 2004, in Tables 2 and 3 are based largely on the three United States climate models that participated in AR4: the National Center for Atmospheric Research Community Climate System Model (CCSM3) (Collins et al., 2006; Barth et al., 2000), the Geophysical Fluid Dynamics Laboratory Coupled Model (CM2.1) (Delworth et al., 2006, Ginoux et al., 2006; Tie et al., 2005; Horowitz, 2005), and the Goddard Institute for Space Studies Model-E (Hansen et al., 2005; Schmidt et al., 2006). Although treatment of aerosols differs somewhat from model to model, for most aerosol properties and processes the differences among models within a given generation of climate models are considerably less than the differences between successive generations.

Most of the important aerosol species (sulfate, organic carbon (OC), black carbon (BC), mineral dust, and sea salt) are treated in climate models for the first time in fourth generation models, and for the first time the sulfate chemistry is embedded in the climate model. However concentrations of most other aerosol species are prescribed from offline simulations with global aerosol models. Inevitably this approach for the other species is a compromise, because it does not account for correlations between climatic variables affecting aerosol influences (such as cloudiness) and the aerosol loading and further because it does not account for aerosol influences on, for example, the hydrological cycle, that can further influence the amount and properties of aerosols. Hydrophilic (soluble) and hydrophobic (insoluble) forms of OC and BC are treated separately. The size distribution (the probability distribution of number with particle size) is prescribed for each aerosol species, so that only the mass concentration of each species is simulated. The aerosol mixing state (which describes whether individual particles are composed of only a single species (external mixing) or of multiple species (internal mixing)) is assumed to be external, so that each aerosol type is assumed to consist of only a single species. This assumption greatly simplifies the representation of aerosol optical properties (absorption coefficient, scattering coefficient, and asymmetry parameter) and their dependence on relative humidity. Optical properties of aerosol particles depend to first order on the refractive index and particle size, and to a lesser extent particle shape. With the assumption of a prescribed size distribution, particle shape and composition for each aerosol type, the refractive index and the optical properties can be prescribed for each aerosol type, but this approach cannot account for variability in these properties for a given aerosol type. Similarly for specified particle composition, particle hygroscopic growth can be prescribed for each aerosol type. The cloud condensation nuclei (CCN) spectrum $N_{CCN}(s)$, which is the concentration of aerosol particles that can nucleate cloud droplets as a function of supersaturation with respect to water, is either ignored or treated using empirical relationships that are suitable at best only for externally-mixed aerosols.

Representation of aerosol processes in fourth-generation climate models is highly simplified. Emissions are prescribed, rather than being treated as dependent on climate model variables such as wind speed and soil moisture. Concentrations of oxidant species OH, O_3, H_2O_2 responsible for conversion of SO_2 and DMS to sulfate are likewise prescribed, typically at monthly mean values, rather than being generated in the model. In most models VOCs are instantaneously oxidized with a uniform prescribed yield. New particle formation and coagulation are not treated as these processes do not affect aerosol properties when the particle size distribution is prescribed. Condensation of the oxidized precursor gases on particles is assumed to occur instantaneously. If OC and BC are treated as distinct hydrophobic and hydrophilic components, aging of

hydrophobic to hydrophilic is treated using a uniform prescribed conversion time. Aging of dust is neglected. Water content of particles is assumed to be governed by thermodynamic equilibrium with relative humidity, with hysteresis (dependence of the hydration state on the history of its environmental relative humidity) neglected in most models by assuming particles are always hydrated at humidities above a specified efflorescence point. Aerosol activation is treated by assuming that a prescribed mass fraction of each aerosol type (typically 100% for sulfate, sea salt and hydrophilic OC and BC, 0% for dust and hydrophobic OC and BC) forms droplets in clouds; with empirical expressions relating cloud drop number concentration to mass concentration. A bulk treatment of aqueous-phase chemistry is used, in which oxidation of dissolved SO_2 in cloud droplets is independent of droplet size but depends on droplet pH, with pH diagnosed assuming a prescribed ratio of ammonia to sulfate; cloud properties for chemistry calculations are provided by the host GCM. Vertical transport and removal of aerosol by cumulus clouds is treated using cumulus parameterizations with poorly constrained estimates of precipitation area. In-cloud and below-cloud scavenging are treated using precipitation rates that are not influenced by the aerosol; treatment of in-cloud scavenging assumes complete of the activated aerosol in clouds.

Aerosols in Fifth Generation Climate Models. The treatment of aerosol properties and processes in fifth generation models will be much more complete than in fourth generation models and increasingly based on understanding of the pertinent processes. More processes will be represented, and in greater detail. With few exceptions, the advances will build on the current generation of global aerosol models (Easter et al., 2004; Tie et al., 2005, Koch et al., 2006). As many aspects of the global aerosol models have already been evaluated in comparisons with observations (Ghan et al., 2001a,b; Easter et al., 2004; Tie et al., 2005; Kinne et al., 2005; Koch et al., 2006; Bates et al., 2006), much of the effort involved in upgrading the aerosol treatment in fifth generation climate models will involve transferring the treatments from the global aerosol models to the global climate models. Within the DOE program this component of the work is carried out largely in the Climate Change Prediction Program. Previous work in the ARM program, which produced treatments of aerosol impacts on clouds (Ghan et al., 1997; Ovtchinnikov and Ghan, 2005; Liu et al., 2005) and parameterizations of aerosol radiative properties (Ghan et al., 2001b), will have an impact on the fifth generation models. The ASP provides field measurements and uses them to evaluate specific processes represented in the models.

Aerosol properties for the fifth generation of climate models will have numerous additional degrees of freedom . New species to be included in the models will include nitrate and ammonia/ammonium, and all aerosol species will be simulated online rather than offline, so that concentrations will vary on hourly rather than monthly timescales and will interact with the meteorology that is simulated in the model. The aerosol size distribution will be calculated rather than prescribed. Several different methods for representing the evolution of aerosol size distribution and composition (modal, quadrature method of moments, and sectional) are available. The simplest, the modal method (Whitby and McMurry, 1997; Easter et al., 2004; Liu et al., 2005), assumes a log-normal size distribution for each aerosol type and calculates its number and mass concentrations from separate conservation equations. The quadrature method of moments (QMOM; McGraw, 1997; Wright et al., 2001; Yoon and McGraw, 2004a,b) is more general in that it does not assume a log-normal size distribution; aerosol properties and their evolution are calculated from the moments of the size distribution by Gaussian quadratures. The

sectional method (Adams and Seinfeld, 2002) is most general as the size distribution is explicitly represented, but it requires extensive computational resources in storing, evolving, and transporting large numbers of variables. For this reason the modal and QMOM methods are most likely candidates for fifth generation models. Representation of the aerosol mixing state will accommodate a combination of external and internal mixing so that aerosol forming by condensation will be capable of being internally mixed with primary particles and/or freshly nucleated particles. Two forms of hydrophobic BC and OC will be treated, one purely hydrophobic, the other internally mixed with sulfate, nitrate, and ammonium. Aging of BC and OC will be expressed in terms of the condensation and coagulation rates, so that fresh BC and OC mass and number are transferred to the internally mixed aerosol. Although treating internal mixing can reduce the number of aerosol types, it complicates the representation of aerosol optical properties, hygroscopicity, and CCN activity because, as is the case with actual ambient aerosols, those properties depend on the now variable composition. Fifth generation models will accommodate internal mixing by using mixing rules for refractive index and hygroscopicity pertinent to particle growth with relative humidity and to CCN activity. Other mixing rules may be used to represent the optical effects of inclusions of insoluble material in particles consisting largely of water-soluble material.

Consistent with the online representation of aerosol properties, emissions of DMS, sea salt, and dust will be calculated online using the simulated winds and (for dust) surface moisture and vegetation cover. Likewise oxidation of aerosol precursor gases will be calculated using oxidant concentrations that are generated online by a gas-phase oxidant chemistry module. VOCs will be separated into multiple classes; the yield of new particulate mass will depend on the total amount of organic aerosol (Chung and Seinfeld, 2002; Tie et al., 2005). New particle formation will be introduced as a source of aerosol number, probably using a parameterization of binary nucleation of water and sulfuric acid vapor (Jaecker-Voirol and Mirabel, 1989; Harrington and Kreidenweiss, 1998; Vehkamäki et al., 2002). Condensation of oxidized precursor gases on existing aerosol particles will be treating using mass transfer theory, so that condensation can be distributed across multiple aerosol types. Coagulation of particles both within each type (which reduces number and increases mean size) and between types will be represented to accurately represent particle number concentration. Dust will age as condensation of sulfuric acid and secondary organic onto dust modes changes the bulk hygroscopicity and refractive index of each mode.

Uptake of water by particles will be represented in terms of the bulk hygroscopicity using Köhler theory, with explicit treatment of hysteresis so that dry and hydrated aerosol states are distinguished. Activation of aerosols to form cloud droplets will be expressed in terms of updraft velocity and the aerosol properties (number, size, and hygroscopicity) for all types (Abdul-Razzak and Ghan, 2000; Fountoukis and Nenes, 2005), so that aerosol indirect effects and the competition between aerosol types can be treated in a physically-based manner. Aqueous chemistry will depend on the pH, calculated from the ratio of sulfate to ammonia, which will be allowed to vary. The cloud fraction will be determined using physically-based parameterizations. Convective transport and removal of aerosol will be improved by using new cumulus parameterizations that diagnose precipitating area and treat cloud microphysics. In-cloud scavenging will use the activated aerosol and treat the dependence of precipitation on the number activated. Below-cloud scavenging will use size-dependent collection efficiencies.

In sum, much more detailed representation of aerosol processes and properties is expected in the fifth generation of climate models than in earlier generations. Representation of each of these processes rests on improved understanding of the processes themselves and on improved ability to efficiently and accurately represent this understanding in models, both of which are expected to be greatly advanced by ongoing research.

Aerosols in Sixth Generation Climate Models. Although it is difficult to anticipate the treatment of aerosol processes in the sixth generation of climate models so far out into the future, it is clear that this treatment will rely on advances in understanding provided by programs such as the ASP and ARM. For example, recent work funded by NASA and NSF has shown that current understanding of the formation of secondary organic aerosol (SOA) leads to substantial underestimates in the simulated concentrations of OC (Heald et al., 2005). Laboratory and field experiments will provide the foundation for a new generation of models of SOA formation. The challenge will be to condense that understanding into process models that are simple enough to be used in global climate simulations but at the same time provide accurate representations of the process.

It is known from field studies that current models based on binary homogeneous nucleation of sulfuric acid and water yield new particle formation rates that are often far smaller that the measured rate (Weber et al., 1999). In such cases new particle formation rates may be better explained by ternary homogeneous nucleation. of sulfuric acid, ammonia, and water (Napari et al., 2002a,b), or of sulfuric acid, organic acid and water (Jimenez et al. 2003; Zhang et al., 2004), or by ion-induced nucleation of sulfuric acid and water (Lee et al., 2003; Lovejoy et al., 2004). Laboratory data for multiple precursor gases can be accurately parameterized by the nucleation theorem (McGraw and Wu, 2003; McGraw, 2005), but further work is needed to account for the influence of background aerosol on the nucleation rate and subsequent particle growth by condensation and coagulation (McMurry et al, 2005). These parameterizations also need to be evaluated and refined using field measurements and, depending on the outcome, incorporated in sixth generation models. It seems clear that treatment of subgrid variability will also be required because the nucleation rate is a highly nonlinear function of the precursor gas concentrations. For these reasons much attention needs to be paid to issues of scale.

Representing aerosol size distributions and size dependent composition is essential. As explicit representation would seem not to be computationally feasible in climate models, alternative approaches must be investigated. Although representation of the aerosol size distribution by the modal approach is capable of efficiently representing multiple aerosol types under many conditions (Whitby et al., 1978), the inherent assumption of a log-normal size distribution for each mode can break down. For example, aerosol activation in cloud updrafts typically separates the size distribution into activated particles and particles too small to be activated. The resultant discontinuities in the size distributions of the activated and unactivated particles are not well approximated by log-normal functions (Zhang et al., 2002). Possible solutions to this problem are provided by the more general and more accurate sectional, QMOM and piecewise log-normal (von Salzen, 2005) treatments, with differing computational burdens. The ASP and CCPP can contribute to comparing and testing these and other potential approaches..

Uptake of water exerts an important and sometimes dominant influence on aerosol optical properties. Although the equilibrium Köhler treatment seems appropriate for submicrometer particles, this approach may not work for larger particles, for which the change in particle size may lag changes in relative humidity. Such kinetic effects might be treated by an explicit dynamic form of the Köhler theory. Although this treatment is straightforward for parcel models in which the time dependence of the ambient relative humidity is known, extending it to the Eulerian framework of climate models will require alternate approaches such as using the turbulence kinetic energy and the vertical gradient of relative humidity.

Evaluation and Improvement. Although process modules in current global aerosol models can be applied to climate models, these modules have not been evaluated sufficiently to allow them to be used with confidence in climate models, and many such modules need to be improved to provide the basis for the treatment of aerosols in future generations of climate models. Ultimately improvement of model based representation of aerosol processes and properties rests on understanding that is grounded in laboratory studies, theory, and field measurements. The approach to developing modules to describe specific aerosol processes is often a cyclical one, involving perhaps initial field observations, such as the widespread occurrence of new particle formation (Komppula, 2003) identification of the chemical and physical conditions under which this occurs, examination in the laboratory, identification of unmet measurement needs, and subsequent field studies to compare calculated and measured rates of new particle formation under a variety of conditions. Once such a module is developed, it must be made suitable for incorporation in large scale aerosol models. This may require considerable effort, both in meeting computational constraints and in accounting for issues such as subgrid variability, which can greatly affect highly nonlinear processes.

Much of the advance in understanding aerosol processes in recent years has derived from improvements in instruments for determination and characterization of aerosols and aerosol precursors. Table 4 lists some key capabilities in measurements of properties of aerosols and, precursor gases (H_2SO_4, NH_3, VOC) and oxidants (OH). New techniques for characterizing aerosol composition can provide information that is highly speciated, quantitative, and size-resolved for ionic species, organics, and refractory materials such as dust. Methods of electron microscopy and microanalysis, which can characterize the size, morphology, composition, phase and internal structure of single particles (Figures 3), can be coupled with time-resolved aerosol collection on substrates to follow temporal variations of specific types of field collected individual particles. Other techniques, such as PILS (Particle into Liquid Sampler), AMS (Aerosol Mass Spectrometry), and SPMS (Single Particle Mass Spectrometry) provide real-time measurements with a time resolution of seconds (Figure 4). Although PILS provides little size-resolution, it provides accurate concentration measurements of soluble ionic aerosol species. AMS provides size-resolution of composition (except for refractory material such as BC, sea salt and mineral dust) for sizes between 30 nm and 2.5 microns. SPMS provides composition information for individual particles as small as 50 nm. Other measurement highlights include aerosol size distribution to diameters as low as 3 nm, water uptake on particles of a selected size as a function of relative humidity, angular distribution of light scattering intensity (Figure 5), and properties of particles that have been scavenged by cloud droplets (by aerodynamically separating them from surrounding gases and unactivated particles; Noone et al., 1988; Twohy et al., 2001, 2005). Thorough reviews of the technology of in-situ aerosol measurement are

provided by McMurry et al. (2000, 2004) and Sullivan and Prather (2005). Further information on AMS and SPMS systems is available at http://cires.colorado.edu/~jjose/ams.html.

Each of these measurements can be used to evaluate different aspects of the treatment of aerosol properties that must be represented in large-scale aerosol models. The ASP conducts field studies designed to obtain such measurements. A key approach to the analysis of field measurements is the so-called "closure experiment" which consists of comparison of measured and modeled quantities, where the modeled quantity is determined using a process model and measurements other quantities as input variables. Several such closure experiments are given in Table 5. An example of such a closure study is shown in Figure 6, which shows several comparisons of CCN measured as a function of supersaturation and calculated based on measurements of particle size distribution and composition (Gasparini et al 2006). Other examples of property closures include mass concentration and concentrations of individual substances; scattering, absorption and extinction coefficients; optical properties, composition, and size distribution, and hygroscopic growth and composition.

The same closure approach can also be used to examine processes, particularly those processes that dominate the relationship between properties that can be measured. For example, under some conditions the aerosol activation process dominates the relationship between subcloud aerosol and activated aerosol. By measuring the updraft velocity and the properties of the subcloud aerosol and the aerosol activated within cloud droplets, understanding of the activation process and representation of that understanding in process models can be tested using the updraft velocity and subcloud properties as input to an activation model and using the activated aerosol measurements to evaluate the activation simulation. Figure 7 illustrates such an evaluation. Similar closure experiments can be applied to new particle formation, water uptake, droplet collision/coalescence, and in-cloud scavenging.

Such closure experiments provide valuable information about the cause of model shortcomings because, unlike evaluations of integrated models, differences between simulations and observations can be attributed to specific processes. In contrast, when large differences are found in comparisons of simulations by integrated models with field measurements, the differences could arise from errors in the representation of any of many different processes or boundary conditions. Closure experiments focus attention on isolated processes, so that the cause of the problem is much more evident.

Evaluations such as these serve two purposes. First, they provide valuable information about the current level of uncertainty in the treatment of individual processes and in the aerosol simulated by integrated models. Second, they identify problems that need to be addressed and suggest directions for future improvements. For example, Wood's (2005) recent evaluation of droplet collision/coalescence schemes identifies weakness in all schemes and suggests how they might be improved.

Increasingly it is becoming appreciated that advances in understanding of aerosol processes from field measurements are much more likely when multiple measurements of aerosol properties are made simultaneously. An example is shown in Figure 8. Physical measurements, of the dependence of critical supersaturation of particles as a function of their diameter, show marked differences above and below a shallow stratus deck in the vicinity of Pt. Reyes, CA. The

above-cloud particles of the same size require a greater supersaturation, and particles at both altitudes require a greater supersaturation than would be expected for particles consisting entirely of inorganic salts; also shown for reference are measurements made in the eastern Caribbean, which are consistent with an inorganic salt composition. Simultaneous measurements of bulk composition show a larger organic fraction above clouds than below. In the absence of the chemical measurements the reasons for the differences in critical supersaturation would not be known; in the absence of the physical measurements the consequences of the differences in composition would not be known.

In some cases results of field studies might suggest a return to laboratory experiments. For example, recent evaluation of organic aerosol modeling (Heald et al., 2005) suggests fundamental problems with current understanding of the production of secondary organic aerosol from oxidation of VOC. As this understanding is based largely on laboratory experiments, it is clear that new laboratory experiments need to be conducted to examine reaction systems that have not been represented in previous models. It is likely that new chemical mechanisms will have to be developed, based on new lab experiments, to account for the underestimated yield and provide the basis for simplified treatment of these processes in global climate models. This new understanding will in turn need to be evaluated by further field measurements.

Regional and global scale aerosol models. Once modules treating a particular aerosol property or process have been developed and evaluated using laboratory and field measurements, the next challenge is to apply them in regional and global models. Regional models provide an important test bed for evaluating an integrated package of process modules by comparing simulations with field measurements. Much of the research in the DOE ASP is focused on this activity. A second important staging ground for such modules is global aerosol models, which are run at grid resolutions comparable to that of global climate models (typically 100 km) but only for a few years rather than hundreds of years. Such short simulations provide a test bed for evaluating aerosol packages at the grid resolution of global climate models without the high cost of running climate simulations to equilibrium. Applying aerosol packages to simulations at 100 km grid resolution introduces issues of scale, because aerosol packages that have been developed in a regional modeling framework with grid resolutions of 1-10 km resolution may not perform well at grid resolutions of 100 km. Subgrid variability in processes at scales between 10 and 100 km may need to be represented if the package is to perform well at 100 km grid resolution. This challenge can be addressed in a regional modeling framework, but the limited spatial degrees of freedom in a regional simulation at 100 km grid resolution may not provide a rigorous test of the treatment of the influence of scale. Global aerosol models can be evaluated against the rich set of global satellite retrievals of a variety of gas, aerosol and cloud properties (Schoeberl et al., 2006; Kaufman et al., 2003; Bellouin et al., 2005; Sekiguchi et al., 2003; Spinhirne et al., 2005). Although intensive field experiments, regular surface-based in situ measurements at NOAA Climate Monitoring and Diagnostics Laboratory stations, twice-weekly in situ measurements above the ARM Southern Great Plains site (http://www.cmdl.noaa.gov/aero/net/iap/index.html), and surface-based aerosol retrievals by the ARM Raman Lidar (Ferrare et al., 2006), AERONET (Holben et al., 1998), MPLNET (Welton et al., 2002), and EARLINET (Bösenberg et al., 2003; Mattis et al., 2004) provide limited horizontal sampling, they do provide accuracy and vertical resolution lacking in satellite retrievals. These datasets have been used successfully in evaluations of global aerosol model simulations of the spatial and temporal distributions of aerosol mass concentration, composition, size distribution, and optical properties (Kinne et al.,

2003; Easter et al., 2004; Liu et al., 2005; Heald et al., 2005; Tie et al., 2005; Kinne et al., 2005; Koch et al., 2006; Bates et al., 2006). Retrievals from the recently launched CloudSat (Stephens et al., 2002) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) (Winker et al., 2004) instruments will systematically provide vertical profiles of cloud and aerosol from satellite, literally adding another dimension to the evaluation of aerosol models.

Application in Global Climate Models. Once aerosol packages have been evaluated in a global aerosol model they are ready for incorporation into global climate models. Such incorporation can be straightforward if the packages have been designed from the outset for ultimate application to global climate models, even more so if the global aerosol model that served as the staging ground is the atmospheric component of a global climate model. However, further work and testing is required if, for example, the climate model resolution differs from that of the aerosol model, or if the treatment of clouds in the global aerosol model differs substantially from the treatment in the climate model. Such differences can have profound impacts on the spatial distributions of aerosol simulated in the global climate model and on the resulting climate influences. Attention must be devoted also to ensure that the global energy balance is maintained. For these reasons, several climate modeling groups are using the atmospheric component of their climate model as the staging ground for the development and evaluation of new aerosol packages.

Integration. Development of new aerosol packages for global climate models involves many steps, from laboratory and field measurements, to process modeling, evaluation, and refinement, to integrated modeling evaluation, and refinement, to scaling to global models, evaluation, and again refinement. Laboratory and field measurements provide the foundation upon which understanding of aerosol properties and processes is based. Process models provide testbeds for evaluating both the full expression of that understanding in detailed models and for simpler representations suitable for application to global models. Integrated aerosol models provide testbeds for treatments of the full set of processes that control aerosol properties. Global aerosol models provide the setting for evaluating treatments of the subgrid variability in aerosol processes for global conditions that can be sampled from instruments on satellite platforms. Global climate models are the ultimate destination of aerosol process and property modules. Although the development process, from lab and field measurements to regional and global aerosol models and ultimately to global climate models, requires much effort, this process is essential to accurate determination of the climate influences of atmospheric aerosols. Integration of all these components is essential to assure that the understanding of processes that is developed in laboratory studies and field measurements is accurately and effectively represented in successive generations of climate models.

Closing Remarks. Global climate models are important tools for understanding climate change and for development of policy regarding future emissions of greenhouse gases and aerosols. As such, these models must include accurate representation of radiative forcing and other influences of anthropogenic tropospheric aerosols that are significant contributors to climate change over the industrial period. Unlike most greenhouse gases, tropospheric aerosols are quite dynamic, undergoing transformations that modify their properties that affect atmospheric radiation, clouds, and the hydrological cycle. The loadings and properties of these aerosols are quite variable spatially and temporally in large part because of their interactions with other elements of the climate system. These interactions go in both directions, aerosols

influencing other climatic variables and other climatic variables influencing the amount and properties of aerosols. For these reasons it is increasingly becoming recognized that accurate determination of aerosol influences on climate requires actively modeling the loading and properties of aerosols in climate models, rather than calculating these quantities in an aerosol model, offline from the climate model. Here a sequential approach has been outlined whereby increasingly detailed representation of aerosol properties and processes will be incorporated into future generations of climate models. Successful implementation of this approach will rely on advances in understanding of these processes in laboratory and field studies, and advances in representing these processes in aerosol models on regional to global scales. This strategy is being implemented by the several programs within the Department of Energy's Climate Change Research Division, and substantial advances in understanding and quantification of these phenomena can be expected over the next several generations of climate models.

Appendix. Acronyms

AERONET	AErosol RObotic NETwork
AMS	Aerosol Mass Spectrometry
ARM	Atmospheric Radiation Measurement http://www.arm.gov/
APS	Aerodynamic Particle Sizer
ARn	n^{th} assessment report of IPCC Working Group 1
ASP	Atmospheric Science Program http://www.asp.bnl.gov/
BC	Black Carbon
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation
CAPS	Cloud Aerosol and Precipitation Spectrometer
CCN	Cloud Condensation Nuclei
CCPP	Climate Change Prediction Program http://www-pcmdi.llnl.gov/ccpp/
CCSM	Community Climate System Model
CIMS	Chemical Ionization Mass Spectrometry
CNC	Condensation nuclei counter
CVI	Counterflow Virtual Impactor
DMA	Differential Mobility Analyzer
DMS	Dimethylsulfide
DOE	Department of Energy
EARLINET	European Aerosol Research Lidar Network
EDX	Energy Dispersive X-ray
GCM	Global Climate Model YES? NOT General Circulation Model?
H-TDMA	Humidified Tandem Differential Mobility Analyzer
IOP	Intensive Operational Period in ARM program
IPCC	Intergovernmental Panel on Climate Change
MPLNET	Micro-pulse Lidar Network
NASA	National Aeronautics and Space Administration
NSF	National Science Foundation
NOAA	National Oceanic and Atmospheric Administration
OC	Organic Carbon
PCASP	Passive Cavity Aerosol Spectrometer Probe
PILS	Particle Into Liquid Sampler
PIXE	Particle Induced X-ray Emission
PTR-ITMS	Proton Transfer Reaction Ion Trap Mass Spectrometer
QMOM	Quadrature Method of Moments
SEM	Scanning Electron Microscopy
SMPS	Scanning Mobility Particle Sizer
SPLAT	Single Particle Laser Ablation Time-of-flight mass spectrometry
SPMS	Single Particle Mass Spectrometry
SOA	Secondary Organic Aerosol
TEM	Transmission Electron Microscope
VOC	Volatile Organic Carbon

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Table 1. Four stages of research and model development necessary to examine aerosol influences on climate.

STAGE	1	2	3	4
ACTIVITY	Process research: Field and Laboratory studies	Development of 0-D models (Modules) representing processes; comparison with process research studies	Incorporation of modules describing aerosol processes in regional to global aerosol models. Production runs. Assessment of accuracy of aerosol models	Incorporation of representation of aerosol processes in climate model; production runs; comparison with observations
OUTCOME	Improved understanding of processes	Modules: Model based representation of understanding	Evaluated aerosol model incorporating processes	Climate relevant runs; assessment of skill of climate model against present and/or prior climate

Table 2. Treatment of aerosol properties in fourth, fifth and sixth generation climate models

	Treatment		
Property	Fourth Generation	Fifth Generation	Sixth Generation
Mass concentration and composition	Sulfate interactive (online) with climate model. Dust, sea salt, hydrophilic and hydrophobic OC and BC prescribed from offline aerosol model simulations	Interactive sulfate, dust, sea salt, hydrophilic and hydrophobic OC, BC, nitrate, ammonia.	Interactive sulfate, dust, sea salt, hydrophilic and hydrophobic OC, BC, nitrate, ammonia.
Size distribution	Prescribed for each aerosol type except dust. Multiple sizes for dust and perhaps sea salt.	Variable for each aerosol type (modal)	Variable (sectional, QMOM, or piecewise log-normal)
Mixing state	External	Internal and external mixtures.	Internal and external mixtures.
Refractive index	Prescribed	Volume average	Volume average. Treatment of inclusions.
Optical properties	Prescribed, for each aerosol type; function of RH	Parameterized in terms of bulk refractive index and wet effective radius.	Parameterized in terms of bulk refractive index and wet effective radius
Hygroscopicity	Prescribed	Volume average	Thermodynamic equilibrium
CCN spectrum	Empirical	Köhler theory for external mixtures of internally-mixed inorganic and soluble organic salts.	As for fifth gen., plus weakly soluble organics and surfactants.

Table 3. Treatment of aerosol processes in fourth, fifth and sixth generation climate models.

	Treatment		
Process	Fourth Generation	Fifth Generation	Sixth Generation
Primary emissions	Prescribed for all species.	Sea salt, dust emissions depend on wind speed in host model; also on soil moisture for dust.	Sea salt, dust emissions depend on wind speed in host model; also on soil moisture for dust.
Precursor emissions	Prescribed for all precursor gases.	DMS emissions depend on wind speed in host model.	DMS emissions depend on wind speed and ocean chemistry in host model.
Oxidation of precursors	Reaction of SO_2, DMS with prescribed oxidant concentrations. Instantaneous oxidation of VOC with prescribed yield.	Reaction of all precursors with oxidants whose concentrations are calculated in the model. Multiple hydrocarbon groups. Dependence of yield on total organic aerosol.	New hydrocarbon treatment.
New particle formation	Neglected.	Binary homogenous nucleation.	Ternary nucleation ammonia, organics
Condensation of oxidized precursor gases	Instantaneous condensation.	Size-dependent mass transfer treatment.	Size-dependent mass transfer treatment.
Coagulation	Neglected.	Brownian coagulation within, between modes.	Brownian coagulation within, between modes.
Evolution of hygroscopicity of BC, OC and dust ("Aging")	Prescribed hydrophobic-to-hydrophilic conversion time for BC and OC. Neglected for dust.	Separate treatment of coagulation and condensation effects for BC and OC, condensation effects for dust.	Separate treatment of coagulation, condensation, surface chemistry effects.
Water uptake	For external mixtures only. No hysteresis in most models. Equilibrium	Internal and external. Hysteresis treated.	Kinetic effects
Aerosol Activation	Prescribed number activated	Maximum supersaturation and number activated parameterized in terms of updraft velocity and external mixtures of internally-mixed inorganic and soluble organic salts.	Kinetic effects. Activation to ice crystals.

Table 3 (continued)

Aqueous phase reactions in clouds	Bulk treatment (same for all cloud droplets). pH dependence for prescribed ratio of ammonia/sulfate; Poorly constrained cumulus cloud fraction	Bulk treatment (same for all cloud droplets). pH dependence for variable ratio of ammonia/sulfate. Physically-based stratiform and cumulus cloud fraction.	Size dependent cloud drop composition. Reactions in hydrated aerosol.
Convective transport and removal	Cumulus parameterization. Poorly constrained precipitating area.	Cumulus parameterization with physically-based precipitating area.	Statistics from embedded cloud models.
In-cloud scavenging	Autoconversion and precipitation rate independent of aerosol. Cloud-borne aerosol equals activated aerosol.	Autoconversion and precipitation rates depend on aerosol. Influence of collision/ coalescence on cloud drop number concentration and cloud-borne aerosol. Subgrid variability in autoconversion.	Statistics from embedded cloud models. with microphysics dependent on aerosol. Scavenging by ice crystals.
Subcloud scavenging	Prescribed scavenging efficiency.	Size-dependent collection efficiency	Aerosol from evaporated raindrops. Precipitation statistics from embedded cloud models.

Table 4. Advanced techniques for characterization of properties of aerosols,precursor gases, and other key quantities

Property	Instrument/technique (reference)
H_2SO_4 gas concentration	CIMS: Eisele and Berresheim (1992)
NH_3 gas concentration	CIMS: Nowak et al. (2002), Fehsenfeld et al. (2002)
OH gas concentration	Laser induced fluorescence: Martinez et al. (2003)
VOC gas concentration	PTR-ITMS: Prazeller et al. (2003)
Concentration of ionic species in aerosols	PILS: Weber et al. (2001) + ion chromatography.
Size-resolved composition	AMS (thermal vaporization mass spectrometry): Jayne et al. (2000)
Single particle composition	SPMS (laser desorption mass spectrometry): Thomson et al. (2000), Su et al. (2004), Zelenyuk and Imre (2005).
Single particles morphology, composition, phase, internal structure, hygroscopicity	TEM, SEM, EDX microanalysis: Buseck and Anderson (1998); Fletcher et al. (2001); De Bock and Van Grieken (1999); Laskin et al. (2006), Twohy et al. (2005). Environmental SEM and TEM: Ebert et al. 2002; Laskin et al. (2006), Wise et al. (2005)
BC concentration	Thermal-optical. Chow et al. (1993)
Dust concentration	PIXE: Johansson et al. (1995)
Ultrafine number concentration	CNC: Stolzenburg and McMurry (1991)
Particle size distribution	SMPS: Wang and Flagan (1990). nanoSMPS: Woo et al. (2001). DMA: Knutson and Whitby (1975). PCASP: Liu and Daum, (2000).APS: Stanier et al. (2004).
Mixing state	H-TDMA: McMurry and Stolzenburg (1989). SPMS. SEM. TEM. scanning transmission X-ray microscopy.
Activated particle size distribution and composition	CVI: Noone et al. (1988) + CCN+DMA + PCASP + AMS +TEM (Twohy et al., 2001, 2005)
Angular distribution of light scattering, size distribution, and real and imaginary components of refractive index	Polar nephelometer: Barkey et al. (2002)
Absorption coefficient	Photoacoustic: Arnott et al. (1999, 2006). Cavity ring-down extinction – nephelometer scattering: Strawa et al. (2003, 2006). Multiangle Absorption Photometry: Petzold et al. (2005)
Scattering coefficient; backscatter fraction; multiple wavelengths; multiple relative humidities	Integrating nephelometer
Extinction coefficient (multiple wavelengths)	Raman lidar, cavity ring-down, vertical derivative of optical depth measured by airborne sun photometer: Schmid et al. (2006)
Hygroscopic growth	H-TDMA: Gasparini et al. (2006a)

Table 4 (continued)

CCN spectrum	CCN Spectrometer: Hudson (1989), Gasparini et al. (2006b)
Droplet size distribution	CAPS: Baumgardner et al. (2002)
Ground-based and satellite-based remote sensing of aerosol optical depth and column properties	AERONET: Holben et al (1998); Dubovik and King (2000); Kaufman et al. (2002)
Ground-based and satellite-based remote sensing of vertical distribution of aerosol properties	Aerosol lidar: Goldsmith et al. (1998); Turner et al. (2001); Müller et al. (2001); Shipley et al. (1983). Satellite: Kaufman et al. (2003); Winker et al. (2004).
Updraft velocity	Gust probe: Lenschow (1986); Conant et al. (2004).

Table 5. Closure experiments on aerosol properties and processes

| Closure Experiment	Measurements	Reference
Aerosol mass concentration	Ions, BC, organic, dust, total mass	Zhang et al. (2005)
Refractive index	Ions, BC, organic, mixing state, refractive index	??
Radiative absorption	BC, dust, aerosol size distribution, mixing state, absorption	??
Radiative scattering	Ions, BC, organic, dust, aerosol size distribution, relative humidity, mixing state, scattering	Clarke et al. (2002)
Radiative extinction	Ions, BC, organic, dust, aerosol size distribution, relative humidity, mixing state, extinction	Wang et al. (2002)
New particle formation	Condensation nuclei concentration, ultrafine size distribution, H_2SO_4, NH_3, organic vapor	McMurry et al. (2005)
CCN concentration	Ions, BC, organic, dust, aerosol size distribution, CCN concentration	Cantrell et al. (2001); Gasparini et al. (2006b); Rissman et al. (2006)
Water uptake	Ions, BC, organic, dust, aerosol size distribution, humidification size factor, extinction factor	Swietlicki et al. (1999); Dick et al. (2000)
Aerosol Activation / Droplet nucleation	CCN spectrum, ions, BC, organic, dust, aerosol size distribution, updraft velocity, droplet number, activated aerosol	Conant et al. (2004); Meskhidze et al. (2005)
Droplet collision/ coalescence	Droplet size distribution, drizzle size distribution	Wood (2005)
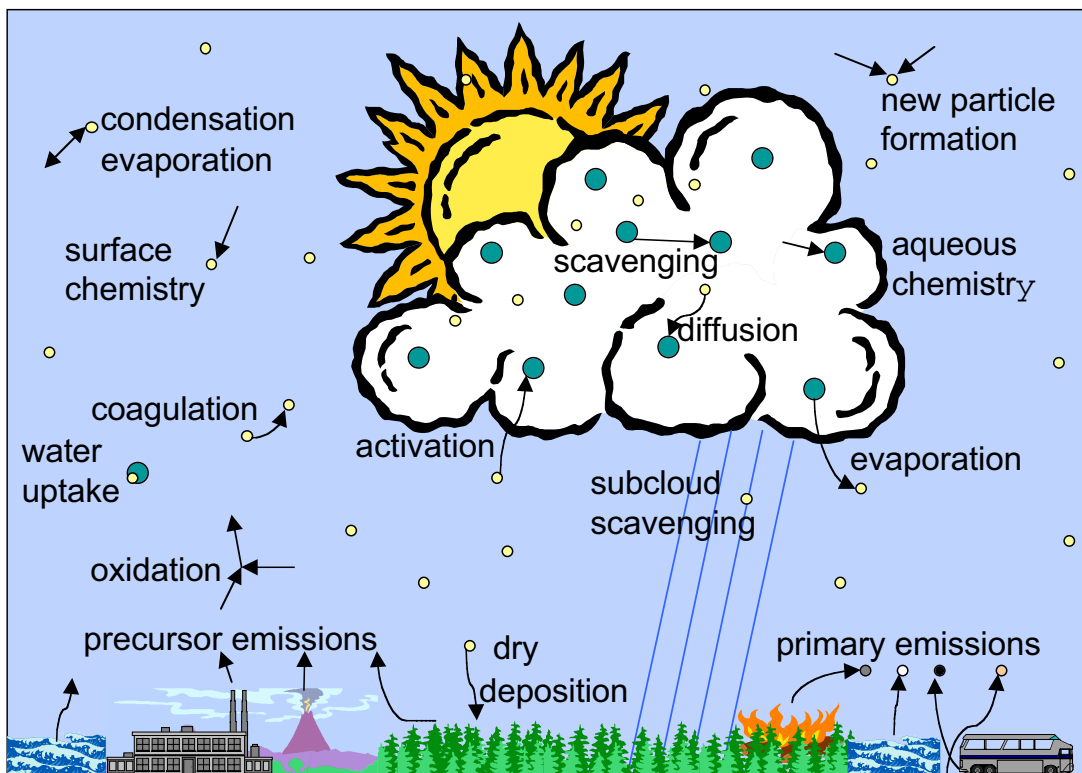
Figure 1. Important aerosol processes that influence climate and which must be accurately represented in future generations of climate models.
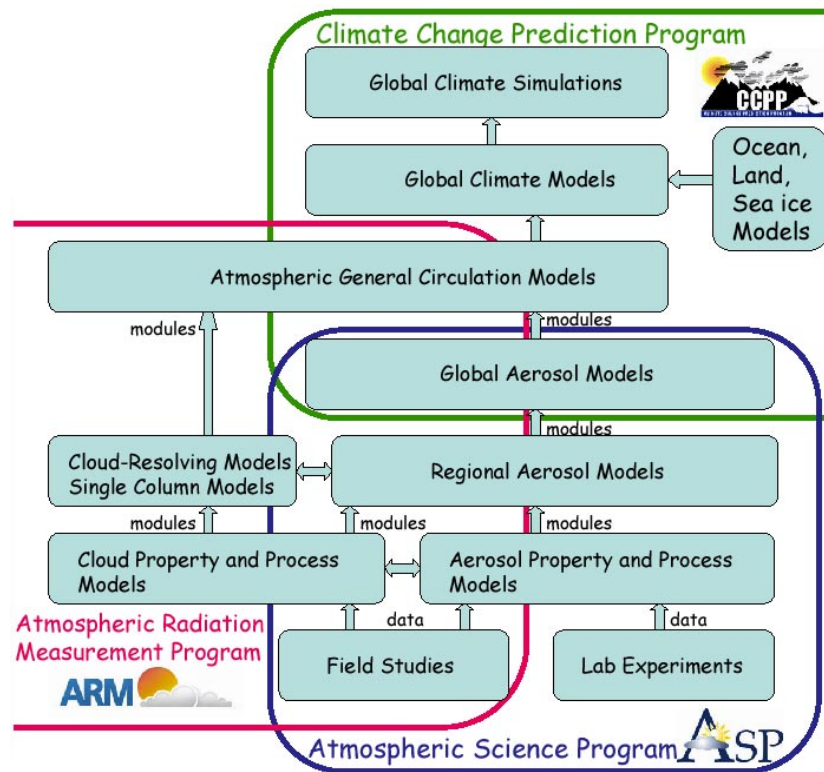
Figure 2. Department of Energy's strategy for improving the treatment of aerosol properties and processes in global climate models. Field studies and laboratory studies provide the foundation. Aerosol property and process models focus on small numbers of aerosol properties or processes. Regional aerosol models represent important aerosol properties and processes by integrating a suite of property and process models for a limited domain and time scales of days. Global aerosol models are like regional aerosol models, but with coarser resolution, a global domain and multiple years of simulation; aerosol processes may or may not feedback on the meteorology. Global atmospheric general circulation models are like global aerosol models, but simulate the meteorology as well as the aerosol. Global climate models are composed of atmosphere models coupled to ocean, land and sea ice models and are typically run for multiple decades to centuries.
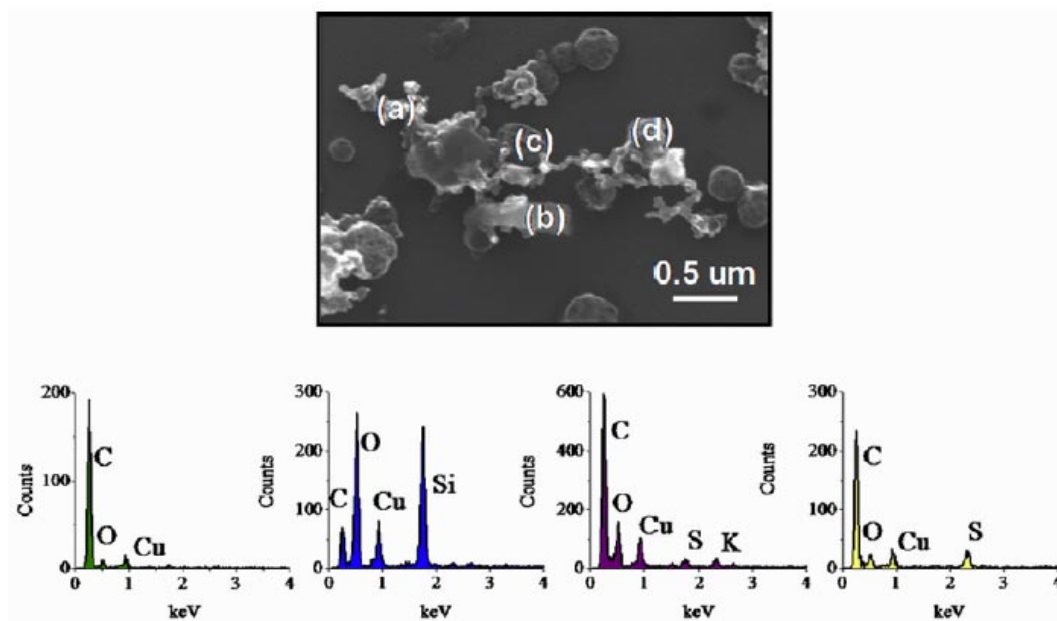
Figure 3. Analysis of a single soot particle collected in Mexico City in 2003 shows complex submicrometer morphology and varying elemental composition as determined by scanning electron microscopy with energy-dispersive x-ray analysis (SEM-EDX). During the residence time of the particle in the atmosphere it has become internally mixed with sulfur, silicon, and potassium; copper is an artifact of the substrate grid. From Johnson et al (2005) Reproduced with permission from Johnson.
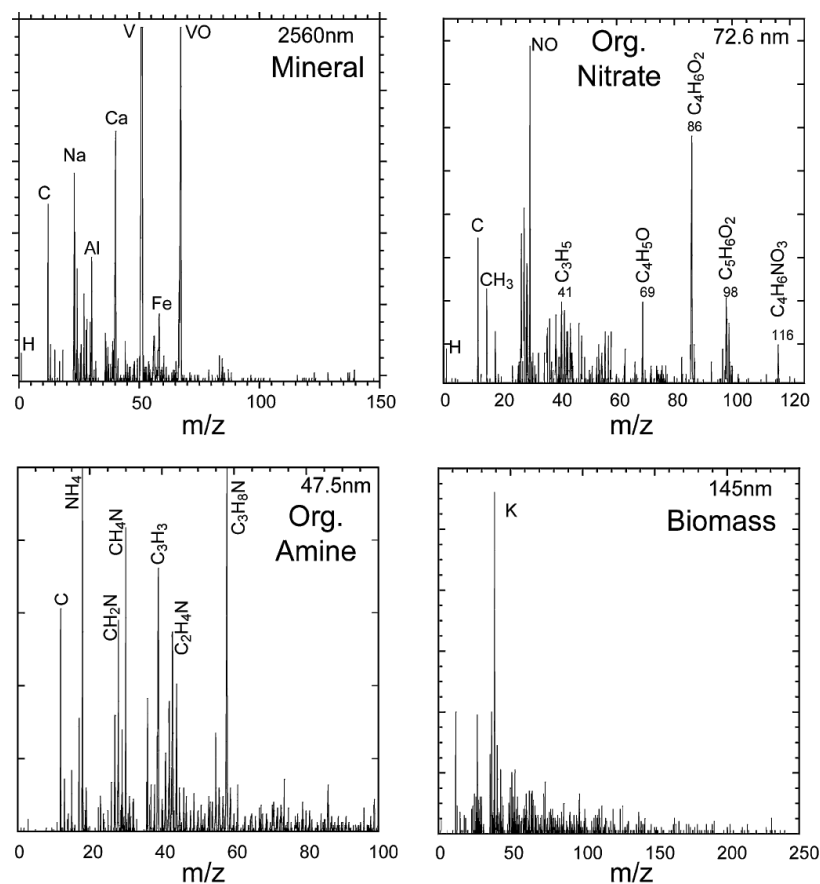
Figure 4. Examples of mass spectra of individual atmospheric aerosol particles sampled by SPLAT (Single Particle Laser Ablation Time-of-flight mass spectrometer; Zelenyuk and Imre, 2005) during a field study in Houston, TX, August, 2000. Indicated size is vacuum aerodynamic diameter From Zelenyuk and Imre (2005). Reproduced with permission from Zelenyuk.
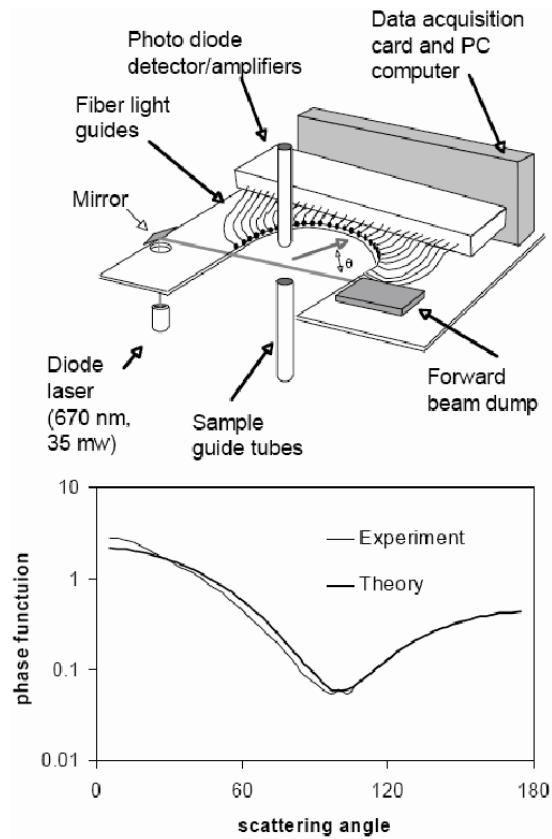
Figure 5. Polar nephelometer to measure angular distribution of light scattering and index of refraction of ambient aerosols. Lower panel shows measured angular distribution of light scattering intensity from laboratory-generated smoke, yielding a size distribution with median diameter 86 nm and variance of 0.113; unpublished results from B. Barkey (UCLA; upper panel modified from Barkey et al., 2002).
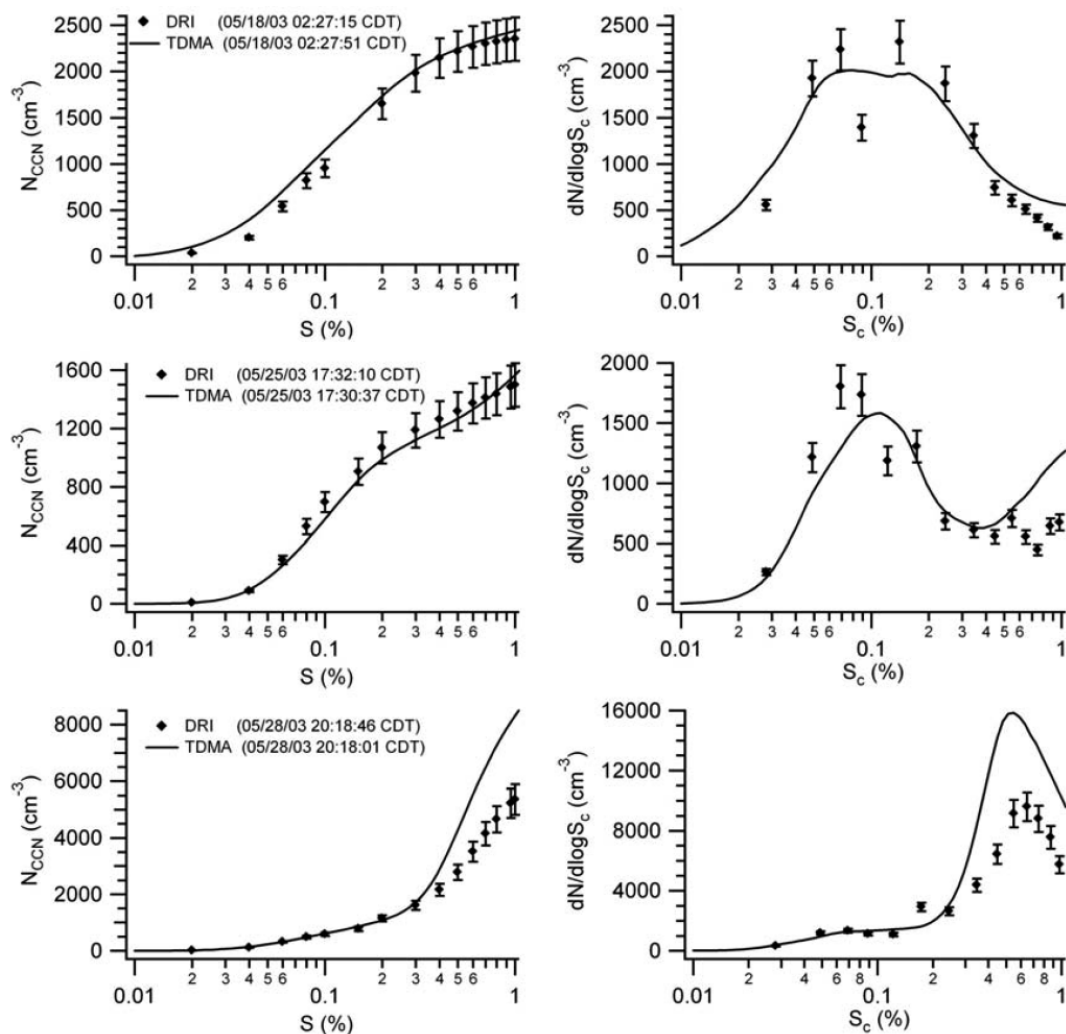
Figure 6. Comparison of cumulative (left) and differential (right) CCN spectra measured during the ARM-ASP Aerosol IOP with the Desert Research Institute CCN spectrometer with spectra calculated from measurements of the size distribution of aerosol number and hygroscopicity. From Gasparini et al (2006), reproduced with the permission of Gasparini..
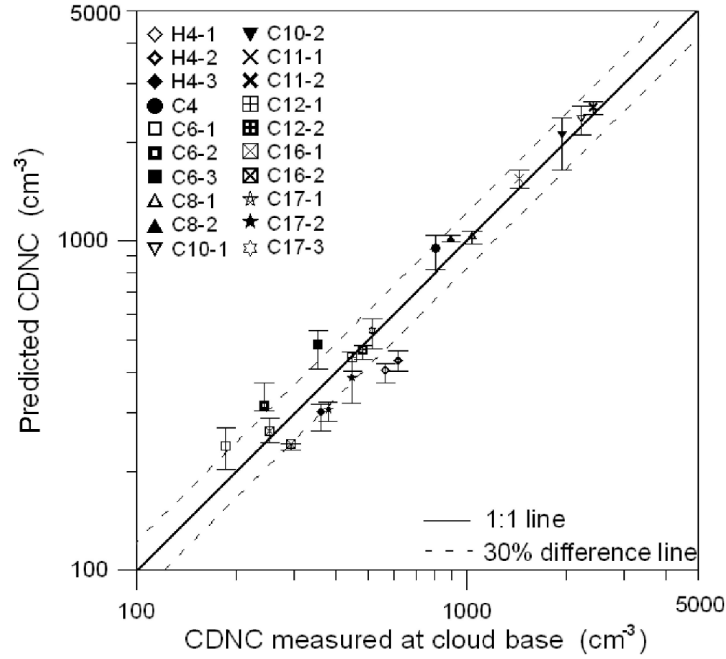
Figure 7. Comparison of droplet number concentration measured on twenty different aircraft flights with that parameterized in terms of measured updraft velocity and measured aerosol size distribution during the CRYSTAL-FACE field experiment (modified from Meskhidze et al., 2005; reproduced with permission of A. Nenes).
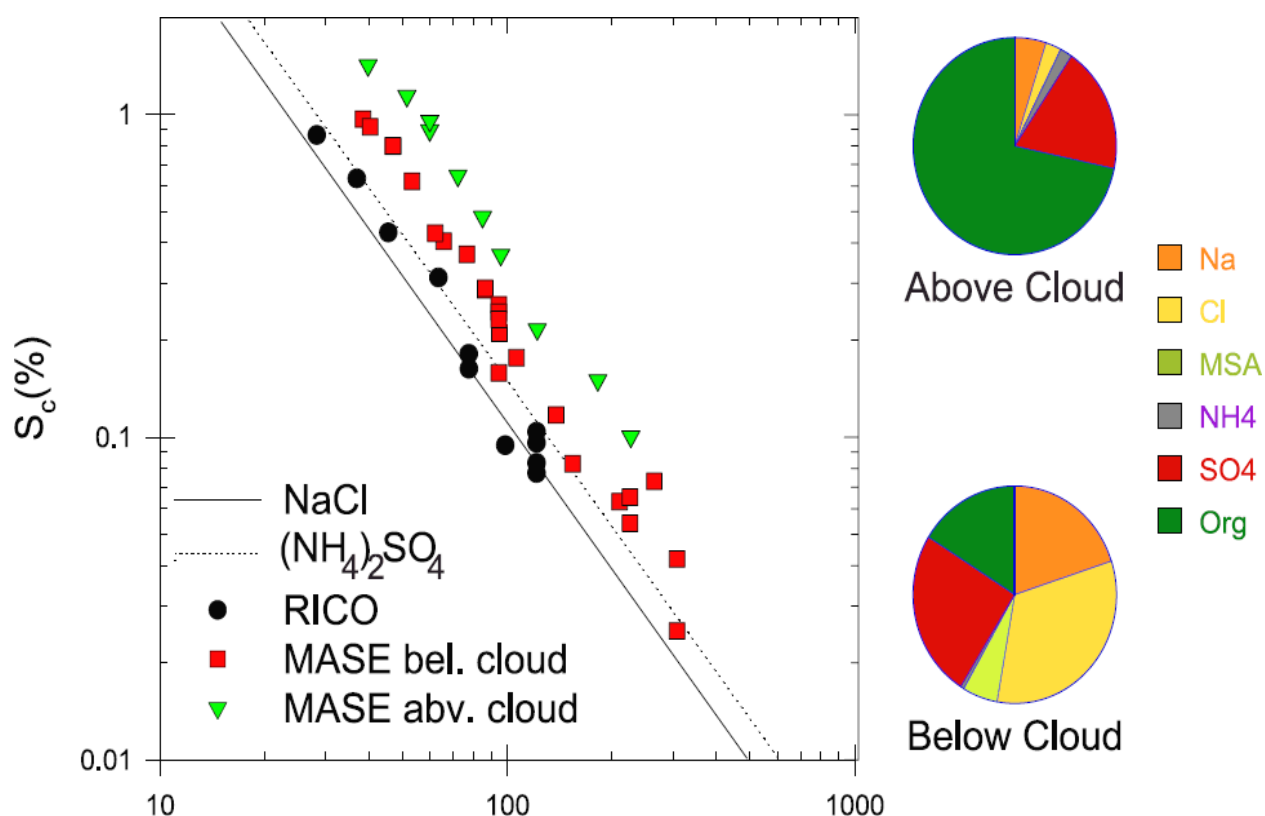
Figure 8. Example of difference in CCN activity of aerosols and relation to composition below (110-170 m) and above (400-470 m) clouds during an Atmospheric Science Program field study MASE (MArine Stratus Experiment) conducted off the coast of California, north of San Francisco, on July 25, 2005. Left panel shows critical supersaturation as a function of particle size from measurements; also shown for comparison are measurements made in clean maritime air in the eastern Caribbean boundary layer (RICO project) and the theoretical dependence for two soluble salts, sodium chloride and ammonium sulfate (J. Hudson, Desert Research Institute, unpublished measurements; Hudson, 1989; Hudson and Da, 1996). Pie charts show ionic composition measured by PILS and organic fraction inferred by difference from total volume, inferred from light scattering at low relative humidity and assumed mass scattering efficiency of 3.3 m^2 g^{-1}; below cloud mass concentration 8.1 ± 0.3 µg m^{-3}; above cloud, 3.8 ± 0.2 µg m^{-3} (Y.-N. Lee, Brookhaven National Laboratory, unpublished measurements).